

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

Appl. No. : 10/706,418 : Confirmation No.: 6797  
Applicant : Rabasco, John J.  
Filed : November 11, 2003  
For : VINYL ACETATE-ETHYLENE CARPET BACKINGS HAVING SPILL  
RESISTANCE  
  
Art Unit : 1771  
Examiner : Salvatore, Lynda  
  
Docket No. : 06426 USA  
Customer No. : 23543

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| <p align="center"><u>Mary E. Bongiorno</u><br/>(Type or print name of person mailing paper)</p> <p align="center"><u>Mary E. Bongiorno</u><br/>Signature of person mailing paper</p> <p align="right"><u>Sept. 12, 2007</u><br/>Date</p> |  |   |

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Alexandria, VA 22313-1450

**APPEAL BRIEF UNDER 37 CFR 41.37(c)**

This appeal is from the final rejection mailed on April 17, 2007.

**REAL PARTY IN INTEREST**

Air Products Polymers, L.P. is the real party in interest in the appeal. The assignment has been recorded at reel 014701/frame 0845.

**RELATED APPEALS AND INTERFERENCES**

There are no related appeals or interferences.

### **STATUS OF CLAIMS**

Claims 1-18 are pending in the application and are the subject of this appeal.

### **STATUS OF AMENDMENTS**

No amendment has been filed subsequent to the Final Rejection of April 17, 2007.

### **SUMMARY OF CLAIMED SUBJECT MATTER**

Applicants have invented an improved spill resistant carpet wherein a spill resistant coating is applied to the underside of the carpet (page 3, [0011].) Spill resistance in a carpet refers to the property of preventing liquids from permeating from the surface to the underside of the carpet and possibly causing damage to the underlying floor (pages 1 and 2, [0004]).

Claim 1 is the sole independent claim and it pertains to a spill resistant carpet wherein a spill resistant coating is applied thereto (page 3, [0011], lines 6-9). The spill resistant coating is comprised of an ethylene-vinyl acetate polymer comprised of crystalline ethylene segments (page 3, [0011], lines 10-11 and page 5, [0021] lines 31-32).

Claim 1 further specifies the ethylene-vinyl acetate polymer is formed by emulsion polymerization of vinyl acetate and ethylene in the presence of a stabilizing system (page 3, [0011], lines 10 and 11). The ethylene-vinyl acetate polymer has a crystalline melting point of from 35-110 °C measured at a heat rate of 20 °C per minute (page 3, [0011], lines 13-14) and a tensile storage modulus of at least  $1 \times 10^5$  dynes/cm<sup>2</sup> at a temperature of 115 °C as measured at 6.28 rad/sec. (page 3, [0011], lines 15-16).

### **GROUND OF REJECTION TO BE REVIEWED ON APPEAL**

Claims 1-18 which are on appeal stand rejected under 35 U.S.C. §103 (a) as being unpatentable over Hoopengardner (US 4,990,399) in view of Kohlhammer et al. (US 6,559,259), and further in view of JP 07195637 A.

### **ARGUMENT**

Following is a summary of the Examiner's position with respect to the obviousness rejection of Claims 1-18, based on the above references:

Hoopengardner is cited as showing a carpet cushion comprised of a compressible foam layer coated on one side with a layer of pressure sensitive adhesive.

Kohlhammer et al. is cited as disclosing water soluble adhesives for textile, non-wovens and compression moulding compositions based upon the polymerization of ethylenically unsaturated monomers with specific showings of vinyl acetate-ethylene polymers. As for the requirements of crystalline melting point and tensile storage modulus, the Examiner presumed the polymers would have such properties (Final Rejection, from page 4, last two paragraphs, to the top of page 5).

JP 07295637-A (JP '637) is cited as disclosing a resin composition comprised of a first and second polymer comprised of a crystalline ethylene-alpha olefin polymer grafted to a carboxylic acid, and thus included within the language of Applicants' claims.

From the above, the Examiner concluded it would have been obvious to employ the adhesive of Kohlhammer et al. comprised of crystalline ethylene segments, as taught by JP '637, in the Hoopengardner carpet (Final Rejection, pages 3 and 4).

Applicant's Response to the Final Rejection of Claims 1-18 Under 35 U.S.C. §103(a)

It is respectfully submitted that the combination of references relied on by the Examiner do not establish a *prima facie* case of obviousness with respect to the claims on appeal. The Examiner has made several technical errors with respect to the teachings in the references, and because of the technical errors, not only would one skilled in the art have failed to produce the spill resistant carpet set forth in Claims 1-18, but such claimed invention would not have been obvious to one skilled in the art.

The first error of the Examiner is in the analysis of Hoopengardner as applied to Applicants' Claim 1. Hoopengardner describes a carpet cushion comprised of a compressible foam layer coated on one side with a layer of pressure sensitive adhesive. A scrim is placed adjacent the pressure sensitive adhesive such that when the carpet is wound upon itself it can be unwound. When the carpet is unwound and the carpet placed on the floor the pressure sensitive adhesive adheres the wall-to-wall carpet to the floor and prevents slipping and movement of the carpet (col. 2, lines 14-19).

Hoopengardner discloses a preference for a hot melt pressure sensitive adhesive to that of a water based pressure sensitive adhesive (col. 4, line 68 to col. 5, lines 1-11). Hoopengardner advises that if a water based pressure sensitive adhesive is employed, alternate means for preventing wicking may be employed, e.g., an impenetrable crust

(sealing film) is applied to the underside of the carpet before the pressure sensitive adhesive is applied (col. 5, lines 13-25).

In terms of Applicants' Claim 1, it can be said that Hoopengardner discloses no more than the preamble of Claim 1, i.e., a spill resistant carpet wherein a spill resistant coating. A hot melt adhesive, which functions as both a pressure sensitive adhesive and spill resistant coating, or a sealing film is applied between the carpet and the water based pressure sensitive adhesive (col. 5, lines 12-25). Hoopengardner does not disclose the use of an ethylene-vinyl acetate polymer. The Examiner has conceded that point (Final Rejection, page 3).

The Examiner has cited the Kohlhammer et al. reference as disclosing ethylene-vinyl acetate/NMA polymers having high mechanical strength and high resistance to water (col. 2, lines 16-20) as a substitute for the adhesive of Hoopengardner. Kohlhammer et al. do not suggest such use, but do list a wide variety of uses such as binders for coatings, plasters, and coverings (col. 9., lines 57-67).

It is submitted that the Examiner's technical position in the application of Kohlhammer et al. for use in the Hoopengardner carpet is without merit and erroneous. If the Examiner is correct that the Kohlhammer et al. polymer is a suitable substitute for the adhesive of Hoopengardner, then the application of 35 U.S.C. §103(a) to the claims on appeal is contradictory. There are two primary reasons. First, in order for the Kohlhammer et al. adhesive to be used as a substitute for the adhesive of Hoopengardner, the adhesive of Kohlhammer et al. must be pressure sensitive. Although many applications are disclosed by Kohlhammer et al., it is apparent that such polymers do not have the property of being pressure sensitive. The polymer films and powder particles may stick together during storage or processing (col. 9, lines 42-46); however, such stickiness does not mean they are pressure sensitive. Comparison Example 7 shows the formation of staple tissues impregnated with various ethylene-vinyl acetate polymers and clearly the polymers do not have the property of being pressure sensitive. If the polymers were pressure sensitive, then nonwovens such as tissues incorporating such polymers would stick to ones hands during use. That would be unacceptable. Therefore, on a strictly technical basis, Kohlhammer et al. is not combinable with Hoopengardner under 35 U.S.C. §103(a).

Second, it has already been established in response to the Office Action of October 4, 2005 that Applicants' claimed ethylene-vinyl acetate polymers having ethylene crystallinity

are not pressure sensitive. (The Examiner cited commonly owned copending applications US 2004/0175589 and 2005/0014013 describing further properties of the claimed polymers. The Examiner noted the ethylene-vinyl acetate polymers were identical to those polymers and these polymers were reported as non blocking.) The fact that Applicants' claimed polymers are not pressure sensitive is an inherent property of these polymers. Assuming *arguendo* that the binders of Kohlhammer et al. were pressure sensitive and therefore combinable under 35 U.S.C. §103(a) for use with Hoopengardner, it follows that it would not have been obvious to substitute Applicants' claimed ethylene-vinyl acetate binders in the Hoopengardner carpet based upon the implied teachings of Kohlhammer et al.

Assuming that the polymers of Kohlhammer et al. are not pressure sensitive then the question under 35 U.S.C. §103(a) turns on whether the properties of the polymers of Kohlhammer et al. are equivalent to those claimed for use in Applicants' carpet and suggested by Kohlhammer et al. for such use. It is respectfully submitted, and has been argued heretofore, that Applicants' claimed ethylene-vinyl acetate polymers have properties which are unexpectedly different from the polymers of Kohlhammer et al. The Examiner has made much of the Kohlhammer et al. disclosure in the Final Rejection at page 3 finding similarity of composition in terms of vinyl acetate-ethylene content, surfactant, etc, and the aqueous polymerization of such monomers. Kohlhammer et al. teach that their polymers have high mechanical strength and water resistance. Given those similarities, the Examiner concluded Applicants' claimed polymers must be identical to those of Kohlhammer et al. However, it is respectfully submitted that conclusion with respect to any perceived process similarities simply is not supported by the teachings in Applicants' specification and by virtue of the declaration of Mr. Daniels filed in the August 2, 2006 response (copy enclosed).

With respect to the Examiner's conclusion that the processes are identical, and therefore the polymers have equivalent properties, that simply is not true. Applicants instruct at page 6, [0026] ways to generate the formation of crystalline ethylene segments in the emulsion polymerization process. They recite the influence of pressure [0027 and 0029], the concentration of thermal initiator [0028], and staged addition of vinyl acetate [0026] as to the final properties of the polymer. Applicants' Example 1 discloses a mode for preparing the polymers and achieving ethylene crystallinity as opposed to the prior art methods of producing polymers having amorphous ethylene segments and no detectable crystalline ethylene segments. Contrast the procedure of Applicants' Example 1 to the processes employed by Kohlhammer et al. in Example 4-6. Applicants use a higher ethylene pressure,

higher temperature and staged addition, just to mention of few of the differences. In the absence of appropriate polymerization conditions, one produces an ethylene-vinyl acetate polymer having amorphous ethylene segments and no detectable crystalline ethylene segments.

To supplement the record and remove any doubt as to property differences between Applicants' polymers and those of Kohlhammer et al., Mr. Daniels carried out an actual run for the purpose of duplicating Example 6 of Kohlhammer et al. and reported the results in his declaration signed on July 28, 2006 and filed on August 2, 2006. The conclusion reached by Mr. Daniels was that the Kohlhammer et al. vinyl acetate-ethylene polymer produced by the Example 6 method did not have detectable crystalline ethylene segments. DSC scans were provided to support the conclusion of Mr. Daniels. Thus, even if were obvious to substitute a copolymer of the type taught by Kohlhammer et al. for the adhesive in Hoopengardner, that would not establish a *prima facie* case of obviousness of the claims on appeal because the carpet would not have been comprised of a spill resistant coating comprised of a polymer formed by emulsion polymerization of vinyl acetate and ethylene and wherein the spill resistant coating of emulsion polymerized ethylene/vinyl acetate had crystalline ethylene segments.

The declaration of Mr. Daniels validates another point with respect to the difference between a spill resistant coating comprised of an emulsion polymerized ethylene-vinyl acetate polymer having crystalline ethylene segments and a storage tensile modulus as claimed vis-à-vis a spill resistant coating comprised of an ethylene-vinyl acetate polymer having amorphous ethylene segments and non detectable ethylene crystalline segments. In sum, Applicants' polymer as claimed offers superior performance when used as a spill resistant coating. Applicants at page 12 [0044] of their specification also illustrate the poor performance of a crosslinked vinyl acetate/ethylene/NMA polymer, which Applicants submit is representative of the polymers described by Kohlhammer et al., i.e., polymers wherein the ethylene segments are amorphous, not crystalline. The polymers of the prior art, such as Kohlhammer et al. may exhibit water and solvent resistance but that does not make them suitable as spill resistant coatings, as the data suggest.

JP '637 (translation supplied) was cited as disclosing a resin comprised of an ethylene-vinyl acetate copolymer having crystalline ethylene segments. In terms of Applicants' polymer as claimed, the document does not support that position. First, there is

no disclosure in JP '637 where such ethylene-vinyl acetate polymers are formed via aqueous emulsion polymerization and have crystalline ethylene segments. JP '637 only reports the first and second resin compositions comprised of ethylene- $\alpha$ -olefin polymers as having ethylene crystalline segments. Such first and second ethylene- $\alpha$ -olefin polymers are formed in the presence of a transition metal catalyst such as a Ziegler-type catalyst (page 4). It is obvious, then, that such first and second resins of JP '637 are not formed in the presence of water. Second, neither component of the second resin composition B comprised of (c) a crystalline ethylene/ $\alpha$ -olefin copolymer and (d) a polymer such as an ethylene/vinyl acetate polymer are formed by emulsion polymerization. In fact there is no disclosure whatsoever in JP '637 regarding the exact nature of the composition of the ethylene/vinyl acetate polymer (d) or how it is prepared. However, one can speculate from the working examples that none of the polymers were prepared by emulsion polymerization given the fact the resulting laminates were formed by first melting the polymers and then molding into a laminate. No one skilled in the art is going to first dry, then, melt an emulsion polymerized ethylene-vinyl acetate polymer for molding a film. In sum, it is submitted that Applicants have shown the numerous technical errors of the Examiner in taking the position that one skilled in the art when combining Kohlhammer et al. with JP '637 in combination with Hoopengardner (see bottom of page 4 of the Final Rejection).

### Conclusion

The Examiner has failed to establish a *prima facie* case of obviousness based upon the references set forth in the record. None of the references disclose an emulsion polymerized ethylene-vinyl acetate polymer prepared in the presence of a stabilizing system wherein a portion of the ethylene is in the form of crystalline ethylene segments having the specified thermal melting point and the required tensile storage modulus. None of the references singularly or in combination show a spill resistant carpet incorporating a spill resistant coating comprised of said polymer or teach anything remotely close within the requirements of 35 U.S.C. §103(a) to Applicants' spill resistant carpet as claimed.

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In view of the foregoing it is requested the Examiner be reversed with respect to the rejection 35 U.S.C. §103(a) with a direction that the application be passed to issue.

Respectfully submitted,

A handwritten signature in cursive script that reads "Mary E. Bongiorno".

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## CLAIMS APPENDIX

What is claimed is:

1. In a spill resistant carpet wherein a spill resistant coating is applied thereto to prevent a liquid spill from permeating through the carpet, the improvement which comprises:

a spill resistant coating comprised of an ethylene-vinyl acetate polymer comprised of crystalline ethylene segments prepared by emulsion polymerizing ethylene and vinyl acetate in the presence of a stabilizing system, said ethylene-vinyl acetate polymer having:

a crystalline melting point ranging from 35 to 110 °C measured at a heat rate of 20 °C per minute; and,

a tensile storage modulus of at least  $1 \times 10^5$  dynes/cm<sup>2</sup> at a temperature of 115 °C and measured at 6.28 rad/sec.

2. The spill resistant carpet of Claim 1 wherein the ethylene/vinyl acetate polymer is comprised of from 15 to 90% by weight of polymerized units of vinyl acetate and from about 10 to 85% by weight of polymerized units of ethylene based upon the total weight of the polymer.

3. The spill resistant carpet of Claim 1 wherein the polymer is comprised of from 25 to 80% by weight of polymerized units of vinyl acetate and from about 20 to 75% by weight of polymerized units of ethylene based upon the total weight of the polymer.

4. The spill resistant carpet of Claim 1 wherein the polymer is comprised of from 35 to 75% by weight of polymerized units of vinyl acetate and from about 25 to 65% by weight of polymerized units of ethylene based upon the total weight of the polymer.

5. The spill resistant carpet of Claim 1 wherein the polymer is emulsion polymerized in the presence of a stabilizing system consisting of a nonionic surfactant and an anionic surfactant.

6. The spill resistant carpet of Claim 1 wherein the polymer has a tensile storage modulus of at least  $2 \times 10^5$  dynes/cm<sup>2</sup> at 115 °C and measured at 6.28 rad/sec.

7. The spill resistant carpet of Claim 1 wherein the heat of fusion of said polymer is from about 5 to 100 joules per gram as measured at a heat rate of 20 °C per minute.

8. The spill resistant carpet of Claim 1 wherein the heat of fusion of said polymer is from about 20 to 50 joules per gram as measured at a heat rate of 20 °C per minute.

9. The spill resistant carpet of Claim 1 wherein said ethylene-vinyl acetate polymer has a glass transition temperature from +25 °C to about -35 °C as measured at a heat rate of 20 °C per minute.

10. The spill resistant carpet of Claim 6 wherein the crystalline thermal melting point of said polymer ranges from 45 to 90 °C as measured at a heat rate of 20 °C per minute.

11. The spill resistant carpet of Claim 1 wherein polymerized carboxylic acid units are present in said polymer in an amount from about 0.2 to about 10% by weight of said polymer.

12. The spill resistant carpet of Claim 11 wherein the polymer has a tensile storage modulus of at least  $2 \times 10^5$  dynes/cm<sup>2</sup> at 115 °C and measured at 6.28 rad/sec.

13. The spill resistant carpet of Claim 11 wherein the polymer is emulsion polymerized in the presence of a stabilizing system consisting of a nonionic surfactant and an anionic surfactant.

14. The spill resistant carpet of Claim 11 wherein the heat of fusion of said polymer is from about 5 to 100 joules per gram as measured at a heat rate of 20 °C per minute.

15. The spill resistant carpet of Claim 11 wherein the heat of fusion of said polymer is from about 20 to 50 joules per gram as measured at a heat rate of 20 °C per minute.

16. The spill resistant carpet of Claim 11 wherein said polymer has a glass transition temperature from +25 °C to about -35 °C as measured at a heat rate of 20 °C per minute.

17. The spill resistant carpet of Claim 12 wherein the crystalline thermal melting point of said polymer ranges from 45 to 90 °C as measured at a heat rate of 20 °C per minute.

18. The spill resistant carpet of Claim 11 wherein the polymer comprises 15 to 90% by weight of polymerized units of vinyl acetate, 10 to 85% by weight of polymerized units of ethylene, and 0.5 to 5% by weight of polymerized units of acrylic acid, based on the total weight of the polymer.

**EVIDENCE APPENDIX**

1. Declaration of Mr. Christian Daniels signed July 28, 2006.
2. Translation of JP 7-195637 A.

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**RELATED PROCEEDINGS APPENDIX**

None.

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

**DECLARATION UNDER 37 CFR 1.132**

Mr. Christian L. Daniels, the declarant, states the following:

1. that he has a Master of Science degree in the field of polymer science;
2. that he is employed by Air Products Polymers, L.P. as a research chemist in the Product Research Group;
3. that he has worked on development of polymer emulsions for use in a variety of products, such as pressure sensitive adhesives, laminating adhesives, nonwoven binders, coatings, caulks, and grouts;
4. that, as a Research Associate, he has considerable experience in the design and development of vinyl acetate based polymer emulsions, including vinyl acetate-ethylene polymer emulsions, for the above applications;
5. that he has carefully reviewed US patent number 5,559,259 B2 (Kohlhammer et al.);
6. that he and Dr. John J. Rabasco, a former Senior Principal Research Chemist at Air Products Polymers, L.P. Product Research Group, supervised the production and analysis of the polymer of Example 6 of US 5,559,259;
7. that Gerhard Koehler, co-inventor of US 5,559,259, gave his approval to the batch sheets prepared by Dr. John J. Rabasco to reproduce the polymer of Example 6;
8. that the polymer of Example 6, US 5,559,259, was prepared at the research facilities of Air Products Polymers, L.P., Allentown, Pennsylvania, according to the following procedure:

Protective Colloid of Example 3, US 5,559,259

The "protective colloid" of example 3, US 5,559,259, was prepared first.

This involved the preparation of an approximately 16% strength 2-acrylamido-2-methylpropanesulfonic acid (AMPS) and N-methylol acrylamide (NMA) copolymer.

The following substances were initially introduced into a 2 liter glass laboratory reactor equipped with a blade stirrer, reflux condenser, jacket cooling and suitable metering

devices: 830.26 g deionized water, 18.06 g of a 50% strength NaOH solution, 8.1 g of acetone, 0.56 g of mercaptopropionic acid (MPA), 74.4 g of solid 2-acrylamido-2-methylpropanesulphonic acid (AMPS), 22.8 g of a 48% strength NMA, and 0.90 g of dodecylmercaptan. The solution was stirred and heated to 80°C. The polymerization was initiated by metering 150 g of a 2.2% strength solution of ammonium persulfate (APS) over a period of 3 hours; 10 minutes after the start of the APS solution a monomer solution and a regulator solution were metered in over a 2 hour period. The monomer solution consisted of: 334.80 g of deionized water, 27.1 g of a 50% strength solution of sodium hydroxide (NaOH), 0.56 g of MPA, 119.87 g of AMPS, and 43.3 g of NMA. The regulator solution consisted of 3.60 g of dodecylmercaptan and 32.50 g of acetone. The reaction was heated to 90 °C after the completion of the monomer and regulator solutions. After the completion of the APS metering solution, the contents were cooled and pH was brought to 7.0 with NaOH. The resulting polymeric solution had a solids content of 16.6%, a pH of 7.0 and a viscosity of 132 centipoise.

#### Ethylene/Vinyl Acetate Polymer Dispersion of Example 6

Preparation of the polymer of example 6, 55% strength ethylene/vinyl acetate polymer dispersion, was accomplished using the protective colloid prepared as described above and a monomer meter solution of 1197.4 g of vinyl acetate and 61.2 g of butyl acrylate. An aqueous metering solution was prepared consisting of: 48.2 g deionized water, 5.58 g of a 50% strength sodium salt of AMPS, 52.0 g of a 40% strength iso-tridecyl alcohol polyglycol ether (15 moles), 10.38 g of acrylic acid, 62.0 g of NMA and 170.6 g of the 16.6% protective colloid solution described above.

The following substances were initially introduced in succession into a 1.05 gallon autoclave equipped with a stirrer, jacket cooling, and suitable metering devices: 1006.2 g of deionized water, 5.24 g of 25% strength sodium vinyl sulfonate, 2.17 g of 30% strength sulfosuccinate half-ester, 8.08 g of 100% strength ethylene oxide/propylene oxide block copolymer, 4.18 g of 48% strength NMA, 13.9 g of butyl acrylate and 119.6 g of vinyl acetate. The contents were then heated to 45°C and ethylene was introduced to pressurize the vessel to 870 psi. The polymerization was initiated by metering a solutions

of 5% strength APS and 2.5% ascorbic acid, both at metering rates of 0.4 g/minute and for a time of 6 hours. The start of polymerization was recognized by a change in reactor temperature, due to the exothermic reaction. Ten minutes after the reaction began, both the monomer and aqueous metering solutions were begun at rates to yield a uniform feed over a period of 4 hours. After the completion of reaction the pressure was let down and the pH was brought to 5. The resultant dispersion had a solids content of 51.0% and viscosity of 1334 centipoise;

9. that the polymer of the polymer dispersion described in #8, above, was analyzed by differential scanning calorimetry (DSC) and displayed an onset glass transition temperature ( $T_g$ ) of  $-0.89^{\circ}\text{C}$ . No crystalline melting point ( $T_m$ ) was detected over the temperature range of  $-65$  to  $+200^{\circ}\text{C}$ , indicating the polymer did not contain any appreciable fraction of crystalline domains (DSC scan is attached);
10. that the DSC scan of the polymer of Example 6 would have been expected by a person of ordinary skill in the art, based on the composition/process conditions disclosed in US 5,559,259;
11. that attached hereto is a DSC scan of the polymer of Example 5 of US Serial No. 10/378,996 (docket # 6120) which is provided as a representative illustration of the differences in thermal properties of polymers having ethylene crystallinity;
12. that these data show that the polymers of US 5,559,259 do not inherently have ethylene crystallinity;
13. that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true and further that these statements are made with knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of title 18 of the United



Declaration under 37 CFR 1.132  
Re: US 5,559,259 (Kohlhammer et al.)  
Page 4

States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.



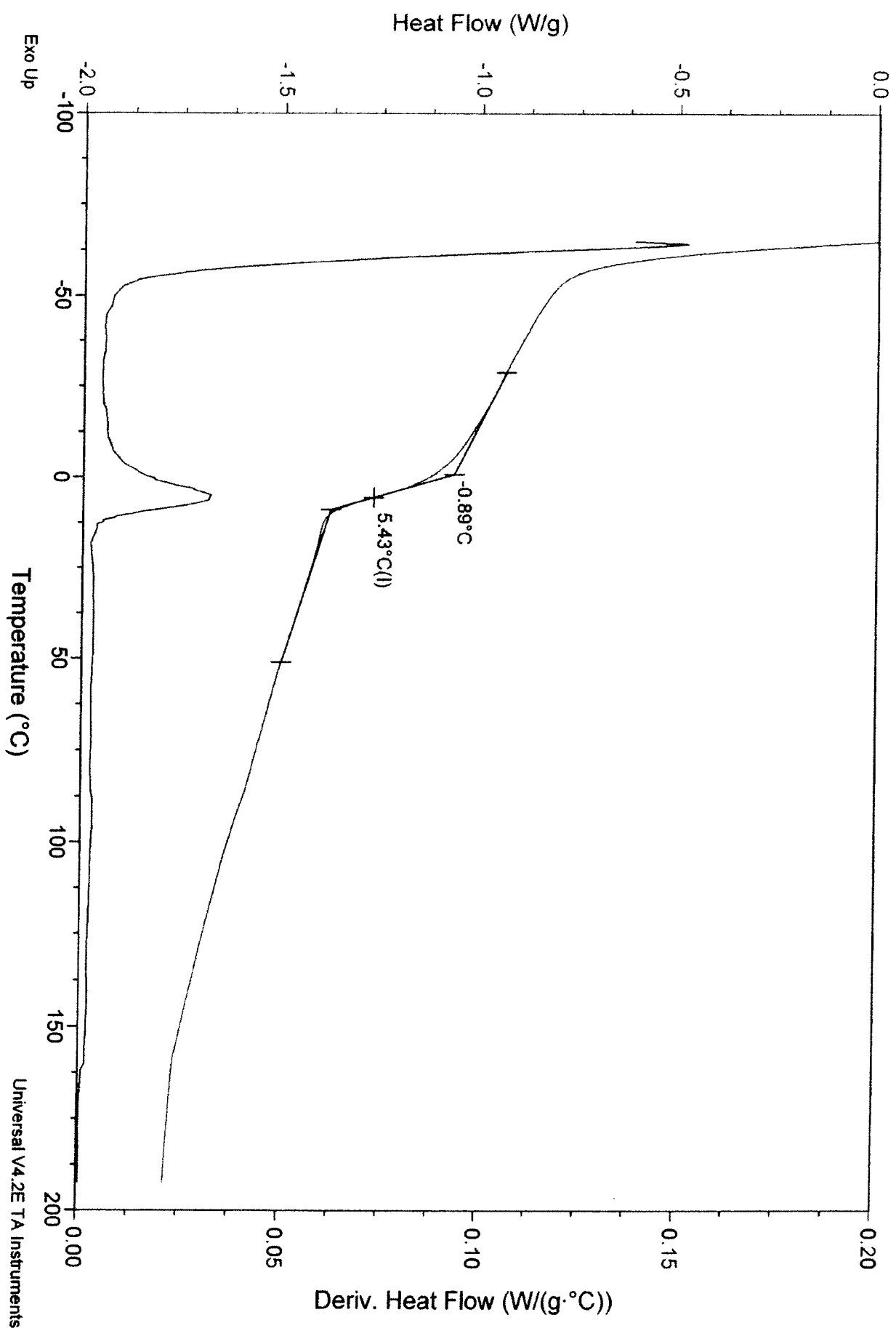
Christian L. Daniels  
Research Associate

24 July 2006  
Date

Sample: EG-242-R3  
Size: 10.0000 mg  
Method: TG(105)(-65)(200)  
Comment: EX#6 US PATENT 6559259

# DSC

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Operator: J COXE  
Run Date: 13-Jul-2006 09:34  
Instrument: 2010 DSC V4.4E

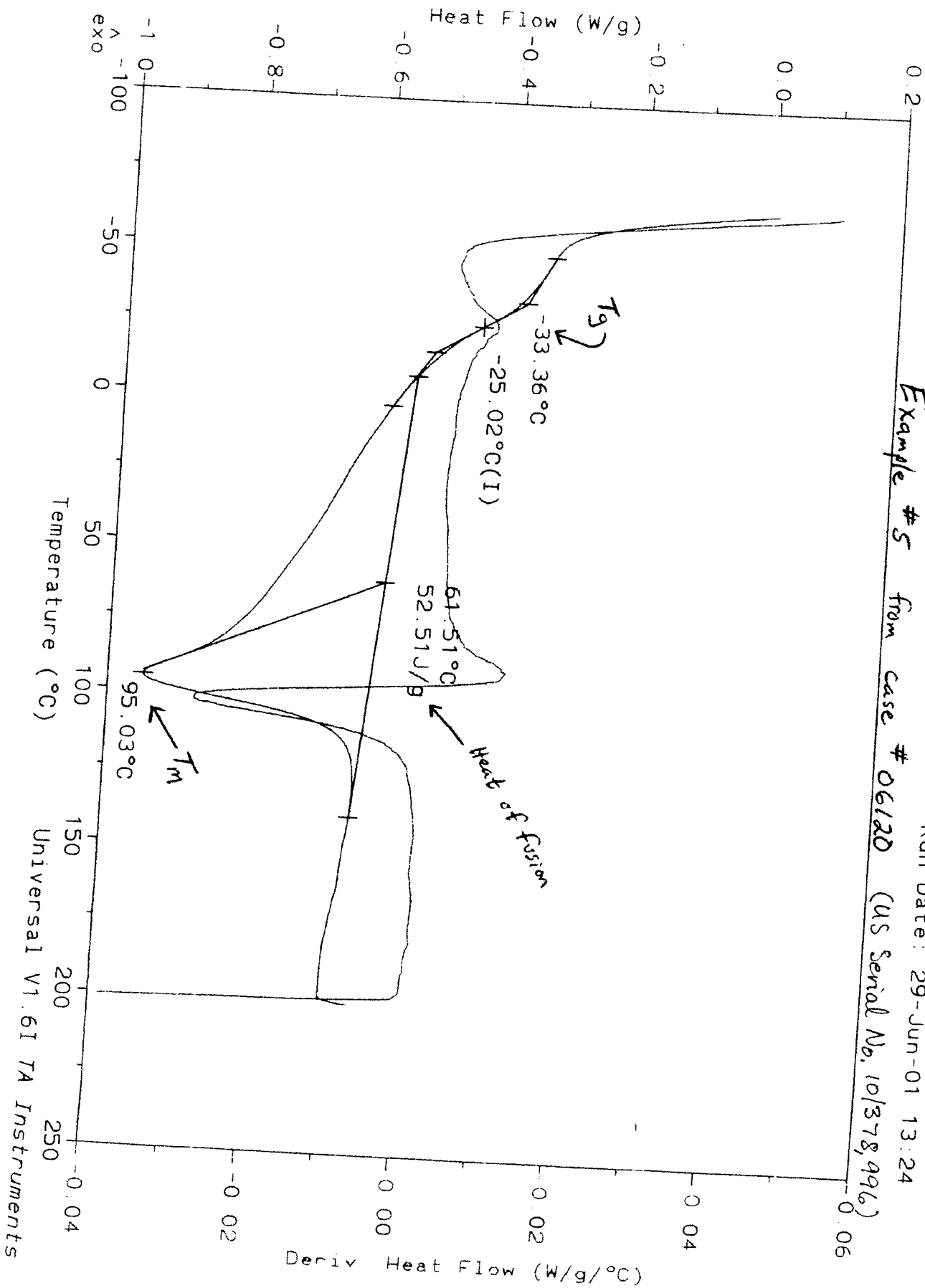


Sample: ZF-461-03  
Size: 32.0000 mg  
Method: TG(100)(-70)(200)  
Comment: JJR

DSC

File: D:\DATA\COXEGD\ZF46103  
Operator: J COXE  
Run Date: 29-Jun-01 13:24

Example #5 from case # 06120 (US Serial No. 10/378,996)



| (51) Int. Cl. <sup>6</sup> | Class. Symbols | Internal Office<br>Registration Nos.: | FI | Technical Classification<br>Field |
|----------------------------|----------------|---------------------------------------|----|-----------------------------------|
| B32B                       | 27/32          | 8115-4F                               |    |                                   |
|                            | 101            | 8115-4F                               |    |                                   |
|                            | 103            | 9268-4F                               |    |                                   |
| 7/04                       |                | 8413-4F                               |    |                                   |
| 27/00                      | C              | 8413-4F                               |    |                                   |
| 27/08                      |                |                                       |    |                                   |

Request for Examination: Not yet submitted

Number of Claims: 2 OL

(Total of 7 pages [in original])

Continued on last page

|                       |              |                 |   |
|-----------------------|--------------|-----------------|---|
| (21) Application No.: | 5-337356     | (71) Applicant: | 000005887                                 |
| (22) Date of Filing:  | Dec 28, 1993 |                 | Mitsui Petrochemical Industries, Ltd.     |
|                       |              |                 | 3-2-5 Kasumigaseki, Chiyoda-ku, Tokyo     |
|                       |              | (72) Inventor:  | Hideshi KAWACHI                           |
|                       |              |                 | c/o Mitsui Petrochemical Industries, Ltd. |
|                       |              |                 | 6-1-2 Waki, Waki-cho, Guwa-gun, Yamaguchi |
|                       |              | (72) Inventor:  | Mineo KUBO                                |
|                       |              |                 | c/o Mitsui Petrochemical Industries, Ltd. |
|                       |              |                 | 6-1-2 Waki, Waki-cho, Guwa-gun, Yamaguchi |
|                       |              | (74) Agent:     | Masataka OSHIMA, Patent Attorney          |

**(54) [Title of the Invention]**

**Resin Laminated Article**

**(57) [Summary] (Amended)**

**[Object]** It is an object of the invention to provide a laminated article with an adhesive layer having exceptional heat resistance, adhesive properties with regard to an ionomer, heat sealing properties, and oxygen barrier properties.

**[Structure]** An ionomer resin laminated article having an adhesive layer made from a resin composition selected from the group consisting of (A) a first resin composition having a density of less than 0.930 g/cm<sup>3</sup> and comprising (a) a crystalline ethylene/ $\alpha$ -olefin copolymer having a density of 0.89-0.94 g/cm<sup>3</sup>, and (b) a non-crystalline or low-crystalline ethylene/ $\alpha$ -olefin copolymer having a density of less than 0.89 g/cm<sup>3</sup> and a degree of crystallinity of 40% or less; (B) a second resin composition comprising (c) a crystalline ethylene/ $\alpha$ -olefin copolymer having

a density of at least  $0.92 \text{ g/cm}^3$ , and (d) an ethylene copolymer selected from the group consisting of an ethylene/vinyl acetate copolymer, an ethylene/acrylic acid copolymer, and an ethylene/ethyl acrylate copolymer; and (C) a third resin composition formed by subjecting the first composition or the second composition to graft modification using an unsaturated carboxylic acid.

**[Claims]**

**[Claim 1]** A resin laminated article comprising (1) an ionomer layer, which has an ionomer, and which is disposed adjacently with regard to (2) a resin composition layer, which has a resin composition selected from a group consisting of (A) a first resin composition, which has a density of less than  $0.930 \text{ g/cm}^3$ , and comprises (a) a crystalline ethylene/ $\alpha$ -olefin copolymer that has been polymerized using a transition metal catalyst and has a density of  $0.89\text{-}0.94 \text{ g/cm}^3$ , and (b) a non-crystalline or low-crystalline ethylene/ $\alpha$ -olefin copolymer having a density of less than  $0.89 \text{ g/cm}^3$  and a degree of crystallinity of 40% or less; (B) a second resin composition, which comprises (c) a crystalline ethylene/ $\alpha$ -olefin copolymer that has been polymerized using a transition metal catalyst and has a density of at least  $0.92 \text{ g/cm}^3$  and (d) an ethylene copolymer selected from the group consisting of an ethylene/vinyl acetate copolymer, an ethylene/acrylic acid copolymer, and an ethylene/ethyl acrylate copolymer; the second resin composition having 95 to 50 wt% of the (c) crystalline ethylene/ $\alpha$ -olefin copolymer and 5 to 50 wt% of the (d) ethylene copolymer based on the total weight of the (c) crystalline ethylene/ $\alpha$ -olefin copolymer and (d) ethylene copolymer; and (C) a third resin composition formed by subjecting the first composition or the second composition to graft modification using an unsaturated carboxylic acid.

**[Claim 2]** A resin laminated article comprising (1) an ionomer layer that has an ionomer, (2) a resin composition layer having a third resin composition, and (3) a resin layer selected from the group consisting of nylon and ethylene /vinyl alcohol copolymer, disposed adjacently in the stated order.

## **[Detailed Description of the Invention]**

**[0001]**

**[Technological Field of the Invention]** The present invention relates to a resin laminated article, and in particular relates to a resin laminated article comprising an ionomer layer and a resin composition layer that has excellent adhesive properties with regard to the ionomer layer.

**[0002]**

**[Prior Art]** Ionomers have excellent heat sealing properties, and so are effectively used in technological fields where such heat sealing properties is required; e.g., as a material for wrapping meat intended for human consumption. However, ionomers cannot be used alone to address the variety of characteristics demanded by their applicable technological fields, and must instead be used in conjunction with other materials; e.g., in the state of a laminated article. An example of a laminated article is known in which an ionomer layer is bonded to an ethylene/vinyl alcohol copolymer layer with a polyethylene adhesive resin layer used as an intermediate layer. However, existing polyethylene adhesive resin layers have drawbacks such as the incidence of interlayer delamination due to inadequate adhesion with the ionomer, and inadequate heat resistance during heat treatments.

**[0003]**

**[Problems to Be Solved by the Invention]** It is an object of the invention to provide a laminated article having a resin composition layer exhibiting exceptional adhesive properties with regard to ionomers, as well as exceptional heat resistance. It is another object of the invention to provide a laminated article with exceptional heat sealing properties, but no incidence of interlayer delamination. It is yet another object of the invention to provide a laminated article having an oxygen barrier property in addition to the exceptional properties as described above. Other objects and benefits of the invention will be made obvious in the following descriptions.

**[0004]**

**[Means Used to Solve the Above-Mentioned Problems]** According to the present invention, the above objects and advantages are achieved by a resin laminated article comprising (1) an ionomer layer, which has an ionomer, and which is disposed adjacently with regard to (2) a resin

composition layer, which has a resin composition selected from a group consisting of (A) a first resin composition, which has a density of less than  $0.930 \text{ g/cm}^3$  and comprises (a) a crystalline ethylene/ $\alpha$ -olefin copolymer that has been polymerized using a transition metal catalyst and has a density of  $0.89\text{-}0.94 \text{ g/cm}^3$ , and (b) a non-crystalline or low-crystalline ethylene/ $\alpha$ -olefin copolymer having a density of less than  $0.89 \text{ g/cm}^3$  and a degree of crystallinity of 40% or less; (B) a second resin composition, which comprises (c) a crystalline ethylene/ $\alpha$ -olefin copolymer that has been polymerized using a transition metal catalyst and has a density of at least  $0.92 \text{ g/cm}^3$  and (d) an ethylene copolymer selected from the group consisting of an ethylene/vinyl acetate copolymer, an ethylene/acrylic acid copolymer, and an ethylene/ethyl acrylate copolymer; the second resin composition having 95 to 50 wt% of the (c) crystalline ethylene/ $\alpha$ -olefin copolymer and 5 to 50 wt% of the (d) ethylene copolymer based on the total weight of the (c) crystalline ethylene/ $\alpha$ -olefin copolymer and (d) ethylene copolymer; and (C) a third resin composition formed by subjecting the first composition or the second composition to graft modification using an unsaturated carboxylic acid.

**[0005]** As described above, the resin laminated article of the invention comprises (1) an ionomer layer, which has an ionomer, and which is disposed adjacently with regard to (2) a resin composition selected from among the first, second, and third resin components.

**[0006]** The ionomer making up the ionomer layer (1) is preferably one in which some of the carboxyl groups of a copolymer comprising ethylene and acrylic acid, methacrylic acid, or another  $\alpha,\beta$ -unsaturated carboxylic acid form a salt with an alkali metal or alkaline earth metal. The  $\alpha,\beta$ -unsaturated carboxylic acid copolymerization ratio is preferably 0.1 to 30 mol%. Salts of sodium, calcium, magnesium, zinc, and other elements can be advantageously used.

**[0007]** The first resin composition for forming the resin composition layer (2) comprises (a) a crystalline ethylene/ $\alpha$ -olefin copolymer that has been polymerized using a transition metal catalyst and has a density of  $0.89\text{-}0.94 \text{ g/cm}^3$ , and (b) a non-crystalline or low-crystalline ethylene/ $\alpha$ -olefin copolymer having a density of less than  $0.89 \text{ g/cm}^3$  and a degree of crystallinity of 40% or less. The transition metal catalyst used here is exemplified by Ziegler-type, Phillips-type, and metallocene-type catalysts.

**[0008]** The (a) crystalline ethylene/ $\alpha$ -olefin copolymer is preferably a random copolymer of ethylene and a  $\text{C}_3$  to  $\text{C}_{20}$   $\alpha$ -olefin. The  $\alpha$ -olefin is preferably a  $\text{C}_4$  to  $\text{C}_{10}$   $\alpha$ -olefin. Examples of  $\alpha$ -olefins preferably include propylene, 1-butene, 1-hexene, 4-methyl-1-pentene, 1-octene, and

1-decene. The (a) crystalline ethylene/ $\alpha$ -olefin copolymer preferably has an ethylene-based polymer unit content of 99 to 80 wt%, and more preferably 99 to 85 wt%. The density of the (a) ethylene/ $\alpha$ -olefin copolymer is 0.89 g/cm<sup>3</sup> or greater and 0.940 g/cm<sup>3</sup> or less, and preferably 0.900 to 0.935 g/cm<sup>3</sup>. The degree of crystallinity of the (a) ethylene/ $\alpha$ -olefin copolymer as measured by X-ray diffraction should be greater than 40%, and is preferably 55 to 80%. The MI value of the (a) ethylene/ $\alpha$ -olefin copolymer is preferably within a range of 0.1 to 30 g/10 min at 190°C.

**[0009]** The (b) non-crystalline or low-crystalline ethylene/ $\alpha$ -olefin copolymer is preferably a random copolymer of ethylene and a C<sub>3</sub> to C<sub>20</sub>  $\alpha$ -olefin. The  $\alpha$ -olefin could be any of those mentioned in the previous description of the ethylene/ $\alpha$ -olefin copolymer. Among those, a C<sub>3</sub> or C<sub>4</sub>  $\alpha$ -olefin is preferred. The (b) ethylene/ $\alpha$ -olefin copolymer preferably has an ethylene-based polymer unit content of 75 to 95 mol%, and more preferably 75 to 90 mol%. The density of the (b) ethylene/ $\alpha$ -olefin copolymer is ordinarily less than 0.89 g/cm<sup>3</sup>, and preferably 0.85 g/cm<sup>3</sup> or greater and less than 0.89 g/cm<sup>3</sup>. The degree of crystallinity of the (b) ethylene/ $\alpha$ -olefin copolymer is 40% or less, and preferably 30% or less. The MI value of the (b) ethylene/ $\alpha$ -olefin copolymer is preferably within a range of 0.1 to 50 g/10 min at 190°C.

**[0010]** With respect to the total weight of the (a) crystalline ethylene/ $\alpha$ -olefin copolymer and (b) non-crystalline or low-crystalline ethylene/ $\alpha$ -olefin copolymer in the first resin composition, the ratio of the (a) ethylene/ $\alpha$ -olefin copolymer is preferably 99-50 wt%, and more preferably, the ratio of the (b) non-crystalline or low-crystalline ethylene/ $\alpha$ -olefin copolymer is 5 to 45 wt%.

**[0011]** The density of the first resin composition is less than 0.930 g/cm<sup>3</sup>, and preferably 0.920 g/cm<sup>3</sup> or less. In addition, the second resin composition that forms the resin composition layer (2) comprises (c) a crystalline ethylene/ $\alpha$ -olefin copolymer that has been polymerized using a transient metal catalyst and has a density of at least 0.920 g/cm<sup>3</sup> and (d) an ethylene copolymer. The (c) crystalline ethylene/ $\alpha$ -olefin copolymer that has a density of at least 0.920 g/cm<sup>3</sup> is preferably a random copolymer of ethylene and a C<sub>3</sub> to C<sub>20</sub>  $\alpha$ -olefin. The  $\alpha$ -olefin could be any of those mentioned in the previous description of the ethylene/ $\alpha$ -olefin copolymer. The (c) ethylene/ $\alpha$ -olefin copolymer preferably has an ethylene-based polymer unit content of 99 to 80 wt%, with the content more preferably being 99 to 85 wt%. The density of the (c) ethylene/ $\alpha$ -olefin copolymer is at least 0.920 g/cm<sup>3</sup>, and preferably 0.920 to 0.935 g/cm<sup>3</sup>. The (d) ethylene copolymer is selected from the group consisting of an ethylene/vinyl acetate



copolymer, an ethylene/acrylic acid copolymer, and an ethylene/ethyl acrylate copolymer. The ethylene/vinyl acetate copolymer preferably has an ethylene-based polymer unit content of 65 to 95 wt%, with the content more preferably being 70 to 95 wt%. The ethylene/acrylic acid copolymer preferably has an ethylene-based polymer unit content of 65 to 95 wt%, with the content more preferably being 70 to 95 wt%. The ethylene/ethyl acrylate copolymer preferably has an ethylene-based polymer unit content of 65 to 95 wt%, with the content more preferably being 70 to 95 wt%. With respect to the total weight of the (c) crystalline ethylene/ $\alpha$ -olefin copolymer having a density of at least 0.92 g/cm<sup>3</sup> and the (d) ethylene copolymer in the second resin composition, the (c) crystalline ethylene/ $\alpha$ -olefin copolymer content is 95 to 50 wt%, and preferably 90 to 60 wt%, whereas the (d) ethylene copolymer content is 5 to 50%, and preferably 10 to 40 wt%. The third resin composition that forms the resin composition layer (2) is obtained by subjecting the first resin composition layer or the second resin composition layer to graft modification using an unsaturated carboxylic acid. The unsaturated carboxylic acid is an unsaturated carboxylic acid or a derivative thereof.

**[0012]** Examples of the unsaturated carboxylic acid or derivative thereof include acrylic acid, maleic acid, fumaric acid, tetrahydrophthalic acid, itaconic acid, citraconic acid, crotonic acid, isocrotonic acid, nadic acid (endo-cis-bicyclo[2.2.1]-hept-5-ene-2,3-dicarboxylic acid), or another unsaturated carboxylic acid; and an acid halide, amide, imide, anhydride, ester, or other derivative thereof. Specific examples of these derivatives include malenyl chloride,<sup>i</sup> maleimide, maleic anhydride, citraconic anhydride, monomethyl maleate, dimethyl maleate, and glycidyl maleate. Of these, an unsaturated dicarboxylic acid or an acid anhydride thereof is particularly suitable, and maleic acid, nadic acid, or an acid anhydride thereof is especially preferable.

**[0013]** Graft modification of the first resin composition and the second resin composition may be performed either directly on the first resin composition or the second resin composition, or performed on all or on some of the components constituting the first resin composition and the second resin composition. Graft modification is preferably performed on the ethylene/ $\alpha$ -olefin copolymers that constitute each of the resin compositions. When an ethylene/ $\alpha$ -olefin copolymer is modified using an unsaturated carboxylic acid or a derivative thereof, the copolymer is preferably graft modified using 0.001 to 15 wt%, and more preferably 0.01-10 wt%, of the

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<sup>i</sup> [Translator's note: This is a possible typographical error for "malonyl chloride."]

unsaturated carboxylic acid or derivative as compared to the ethylene/ $\alpha$ -olefin copolymer before modification.

**[0014]** It is possible that the article having a laminated structure of the invention may have other layers in addition to the ionomer and resin composition layers as described above. Polyethylene, ethylene/vinyl alcohol copolymer, polyester, and polyamide (nylon) are examples of materials that may be used in other layers. These other layers will preferably be disposed adjacently with regard to the resin composition layer. If the other layer is composed of an ethylene/vinyl alcohol copolymer, then the resin component layer preferably comprises the third resin component layer as described above.

**[0015]** In a preferred embodiment of the resin-structured article of the present invention, resin layers selected from the group consisting of (1) an ionomer layer comprising an ionomer, (2) a resin composition layer comprising the third resin composition, and (3) a nylon and ethylene/vinyl alcohol copolymer are disposed adjacently in the stated order.

**[0016]** The laminated article of the present invention can be manufactured by fusing the resin composition or resins constituting the layers, and then laminating them in a molten state. The laminated construction can be molded into the desired configuration by extrusion, casting, inflation molding,<sup>ii</sup> or another molding method.

**[0017]**

**[Working Examples]** The present invention will be described in further detail using working examples, but the present invention is in no way limited by these working examples.

**[0018]** Working Example 1

90 weight parts of an ethylene/4-methyl-1-pentene copolymer that was polymerized using a Ziegler-type catalyst and had a density of 0.920 g/cm<sup>3</sup> (MI = 2.0 g/10 min at 190°C; "LL-1" hereunder) and 10 weight parts of an ethylene/propylene copolymer having a density of 0.88 g/cm<sup>3</sup> (MI=1.1 g/10 min, ethylene content: 80 mol%; degree of crystallinity: 0%; "EPR-1" hereunder) were melt-mixed at 210°C in a single-screw extruder, resulting in an adhesive resin composition. The adhesive composition had a density of 0.916 g/cm<sup>3</sup>, and an MI value of 1.80 g/10 minutes (190°C). An ionomer (Himilan 1601; Na type; manufactured by Mitsui DuPont Polychemical Co., Ltd.), the above-mentioned adhesive resin composition, and a low-

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<sup>ii</sup> [Translator's note: This may be referring to the so-called "tubular film process."]

density polyethylene (Mirason F967; manufactured by Mitsui Petrochemical Industries, Ltd.) were prepared, and each was then melted in the stated order at 200°C, 210°C, and 210°C respectively. A three-layer laminated film (ionomer layer (40 µm)/adhesive resin composition layer (10 µm)/low-density-polyethylene layer (40 µm)) was manufactured using an air-cooled inflation method at a molding rate of 20 m/min and blow-up ratio of 1.0. The adhesive strength between the ionomer layer and the adhesive resin composition layer in the resulting laminated film was 790 g/15 mm.

**[0019] Working Examples 2 through 6 and Comparative Examples 1 and 2**

Three-layer laminated films were formed in same manner as described in Working Example 1 using air-cooled inflation, with the exception that the components used in the adhesive resin composition in Working Example 1 were substituted for those shown in Table 1. The results are shown in Table 1 alongside those of Working Example 1.

**[0020]**

[Table 1]

Table 1

| Working Example                 |                              | 1                   | 2                   | 3                   | 4                   | 5                   | 6                   | Comp.<br>Ex. 1 | Comp.<br>Ex. 2      |
|---------------------------------|------------------------------|---------------------|---------------------|---------------------|---------------------|---------------------|---------------------|----------------|---------------------|
| Adhesive resin<br>composition   | Composition/<br>weight parts | LL-1 90<br>EPR-1 10 | LL-1 80<br>EPR-1 20 | LL-1 70<br>EPR-1 30 | LL-1 90<br>EPR-1 10 | LL-1 80<br>EPR-1 20 | LL-1 70<br>EPR-1 30 | LL-1 100       | PE-1 80<br>EPR-1 20 |
|                                 | MI 190°C<br>(g/10 min)       | 1.80                | 1.70                | 1.55                | 2.10                | 2.25                | 2.40                | 2.00           | 1.90                |
|                                 | Density (g/cm <sup>3</sup> ) | 0.916               | 0.911               | 0.906               | 0.919               | 0.915               | 0.912               | 0.920          | 0.912               |
| Adhesive strength<br>(g /15 mm) |                              | 790                 | 800                 | 870                 | 740                 | 800                 | 830                 | 210            | 460                 |

Notes: LL1: ethylene/4-methyl-1-pentene copolymer  
(MI = 2.0 g/10 min; density: 0.920 g/cm<sup>3</sup>)  
EPR-1: ethylene/propylene copolymer  
(MI = 1.0 g/10 min; density: 0.88 g/cm<sup>3</sup>; ethylene content: 80 mol%)  
EBR-1: ethylene/1-butene copolymer  
(MI = 3.6 g/10 min; density: 0.885 g/cm<sup>3</sup>; ethylene content: 90 mol%)  
PE-1: High-pressure process low-density polyethylene  
(MI = 3.6 g/10 min; density: 0.923 g/cm<sup>3</sup>)

**[0021] Working Examples 7 through 11**

Three-layer films were formed in same manner as described in Working Example 1 using air-cooled inflation, with the exception that the components used in the adhesive resin

composition in Working Example 1 were substituted for those shown in Table 2. The results are displayed in Table 2.

[0022]

[Table 2]

Table 2

| Working Example                |                              | 7                   | 8                   | 9                   | 10                              | 11                  |
|--------------------------------|------------------------------|---------------------|---------------------|---------------------|---------------------------------|---------------------|
| Adhesive resin composition     | Composition/<br>weight parts | LL-1 90<br>EVA-1 10 | LL-1 80<br>EVA-1 20 | LL-1 70<br>EVA-1 30 | LL-1 80<br>EPR-1 10<br>EVA-1 10 | LL-1 80<br>EEA-1 20 |
|                                | MI 190°C (g/10 min)          | 2.00                | 2.00                | 2.00                | 1.85                            | 2.20                |
|                                | Density (g/cm <sup>3</sup> ) | 0.924               | 0.927               | 0.929               | 0.919                           | 0.922               |
| Adhesive strength<br>(g/15 mm) |                              | 770                 | 760                 | 720                 | 750                             | 780                 |

Notes: LL1: ethylene/4-methyl-1-pentene copolymer  
(MI = 2.0 g/10 min; density: 0.920 g/cm<sup>3</sup>)  
EVA-1: ethylene/vinyl acetate copolymer  
(MI = 2 g/10 min; density: 0.95 g/cm<sup>3</sup>; vinyl acetate content: 25 wt%)  
EEA-1: ethylene/ethyl acrylate copolymer  
(MI = 5 g/10 min, ethyl acrylate content: 25 wt%)  
EPR-1: ethylene/propylene copolymer  
(MI = 1.0 g/10 min, density: 0.88 g/cm<sup>3</sup>, ethylene content: 80 mol %)

### [0023] Working Example 12

82 weight parts of an ethylene/1-butene copolymer polymerized using a Ziegler-type catalyst and having a density of 0.923 g/cm<sup>3</sup> (MI = 2.0 g/10 min at 190°C; "LL-2" hereunder), 15 weight parts of an ethylene/propylene copolymer having a density of 0.88 g/cm<sup>3</sup> (MI=2.9 g/10 min; ethylene content: 80 mol %; degree of crystallinity: 0%; "EPR-2" hereunder), and a maleic acid graft-modified polyolefin (maleic acid graft ratio: 2 wt%; "MAHPE" hereunder) were melt-mixed at 210°C in a single-screw extruder, resulting in an adhesive resin composition. The density of this adhesive composition was 0.916 g/cm<sup>3</sup>. An ionomer (Himilan 1601; Na type; manufactured by Mitsui DuPont Polychemical Co., Ltd.), the above-mentioned adhesive resin composition, and a nylon (Amilan CM1021XF; manufactured by Toray Industries, Inc.) were each melted in the stated order at 200°C, 210°C, and 260°C respectively, and a three-layer laminated film (ionomer layer (40 μm)/adhesive resin composition layer (10 μm)/nylon layer (20 μm)) was obtained using an air-cooled inflation method at a molding rate of 20 m/min and a blow-up ratio of 1.0. The adhesive strength in the resulting laminated film was

700 g/15 mm at the interface between the ionomer layer and the adhesive resin composition layer, and delamination was impossible between the nylon layer and the adhesive resin composition layer. Furthermore, an ionomer (Himilan 1605; Zn type; manufactured by Mitsui DuPont Polychemical Co., Ltd.), the adhesive resin composition described above, and an ethylene/vinyl alcohol copolymer (Eval EP-F101A; manufactured by Kuraray Co., Ltd.) were each melted in the stated order at 200°C, 210°C, and 210°C respectively; and a three-layer laminated film (ionomer layer (40 µm)/adhesive resin composition layer (10 µm)/ethylene-vinyl alcohol copolymer layer (20 µm)) was obtained using an air-cooled inflation method at a molding rate of 20 m/min and a blow-up ratio of 1.0. In the resulting laminated film, the adhesive strength at the interface between the ionomer layer and the adhesive resin composition layer was 770 g/15 mm, and the adhesive strength at the interface between the ethylene/vinyl alcohol copolymer layer and the adhesive resin composition layer was 500 g/15 mm.

**[0024]** Working Examples 13 through 15 and Comparative Examples 3 and 4

Three-layer films were formed in same manner as described in Working Example 1 using air-cooled inflation, with the exception that the components used in the adhesive resin composition in Working Example 12 were substituted for those shown in Table 3. The results are displayed in Table 3.

[0025]

[Table 3]

Table 3

| Working Example             |                              | 12                             | 13                             | 14                             | 15                             | Comparative Example 3 | Comparative Example 4          |
|-----------------------------|------------------------------|--------------------------------|--------------------------------|--------------------------------|--------------------------------|-----------------------|--------------------------------|
| Adhesive resin composition  | Composition, weight parts    | LL-2 82<br>EPR-2 15<br>MAHPE 3 | LL-2 80<br>EPR-2 15<br>MAHPE 5 | LL-2 65<br>EPR-2 30<br>MAHPE 5 | LL-2 52<br>EPR-2 45<br>MAHPE 3 | HDPE 98<br>MAHPE 2    | LDPE 80<br>EPR-1 15<br>MAHPE 5 |
|                             | MI 190°C (g/10 min)          | 2.0                            | 2.0                            | 2.1                            | 2.2                            | 1.0                   | 1.5                            |
|                             | Density (g/cm <sup>3</sup> ) | 0.916                          | 0.916                          | 0.908                          | 0.900                          | 0.966                 | 0.912                          |
| Adhesive strength (g/15 mm) | Himilan 1601                 | 700                            | 720                            | 710                            | 710                            | 40                    | 320                            |
|                             | Nylon                        | Delamination impossible        | Delamination impossible        | Delamination impossible        | Delamination impossible        | 800                   | 840                            |
|                             | Himilan 1605                 | 770                            | 780                            | 770                            | 760                            | 50                    | 500                            |
|                             | EVOH                         | 500                            | 650                            | 780                            | 680                            | 20                    | 700                            |

Notes: LL2: ethylene/1-butene copolymer  
(MI = 2.0 g/10 min; density: 0.923 g/cm<sup>3</sup>)  
EPR-2: ethylene/propylene copolymer  
(MI = 2.9 g/10 min; density: 0.88 g/cm<sup>3</sup>; ethylene content: 80 mol %)  
MAHPE: maleic acid graft-modified polyethylene  
(MI = 4 g/10 min, maleic acid graft ratio: 2 wt%)  
HDPE-1: high density polyethylene  
(MI = 1 g/10 min; density: 0.966 g / 15 mm<sup>iii</sup>)  
PE-2: High-pressure process low-density polyethylene  
(MI = 2 g/10 min, density: 0.920 g/cm<sup>3</sup>)  
Nylon: Amilan CM1021XF; manufactured by Toray Industries, Inc.  
EVOH: Eval EP-F101A, manufactured by Kuraray Co., Ltd.

[0026]

(Continued from front page)

| (51) Int. Cl. <sup>7</sup> |       | Class. Symbols | Internal Office Registration Nos.: | FI | Technical Classification Field |
|----------------------------|-------|----------------|------------------------------------|----|--------------------------------|
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|                            | 23/26 | LDP            |                                    |    |                                |
|                            | 51/06 | LLE            |                                    |    |                                |

<sup>iii</sup> Translator's note: Possible typo for "g/cm<sup>3</sup>"